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STUDY OF THE FEASIBILITY OF MEASURING ^{85}Kr THROUGH A NATIONAL SURVEILLANCE SYSTEM

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Abstract

STUDY OF THE FEASIBILITY OF MEASURING ^{85}Kr THROUGH A NATIONAL SURVEILLANCE SYSTEM.

In support of the Environmental Protection Agency's mission of evaluating environmental insult, the Agency's Eastern Environmental Radiation Laboratory has begun to evaluate the feasibility of determining the rate of increase of ^{85}Kr in environmental air caused by the discharge of the gaseous waste product from nuclear facilities. Compressed air is acquired from commercial suppliers in widely distributed locations throughout the United States of America. Krypton is separated from the air and counted by liquid scintillation using the method reported by Cummings et al. Comparison is made with previous determinations as reported by Ehhalt et al. and predictions of Coleman and Liberace. Concentration versus time relations are presented to show any increased levels as more nuclear facilities go into operation. Sampling locations are selected to allow comparison between areas near operating facilities and unaffected areas. The results are analysed to determine if statistically significant differences in levels exist between sampling locations. Variations and concentrations at a given location are also evaluated. Public exposure doses are calculated so as to demonstrate a correlated significance with other environmental radiation dose sources. Comments are offered relative to the future significance of ^{85}Kr in environmental radiation with projected growth in the nuclear industry. Based on the above study, the feasibility of operating a national network is presented, including recommended number of stations, mode of sampling and sampling site selection.

1. INTRODUCTION

Numerous investigators have detected krypton-85 in the atmosphere and have subsequently attributed its existence predominantly to nuclear reactors and nuclear fuel reprocessors [1, 2, 3, 4, 5]. Coleman and Liberace [6] developed a projection of the atmospheric concentrations of krypton-85 which indicated the mean world population dose from this noble gas could reach 50 mrad per year by the year 2060 and in the northern hemisphere population doses by then could exceed 100 mrad per year.

In support of the U. S. Environmental Protection Agency's mission of evaluating environmental insults from industrial pollution sources, the Agency's Eastern Environmental Radiation Laboratory (formerly the U. S. Public Health Service, Southeastern Radiological Health Laboratory) at Montgomery, Alabama, has evaluated the feasibility of establishing a national network to periodically measure environmental concentrations of ^{85}Kr over the continental U. S.

2. OPERATIONAL REQUIREMENTS

In order to develop a functional surveillance system for monitoring ^{85}Kr concentrations, at least four major requirements were considered:

First, an analytical capability was necessary which could measure ^{85}Kr from ambient air with an error of less than 10 percent. This

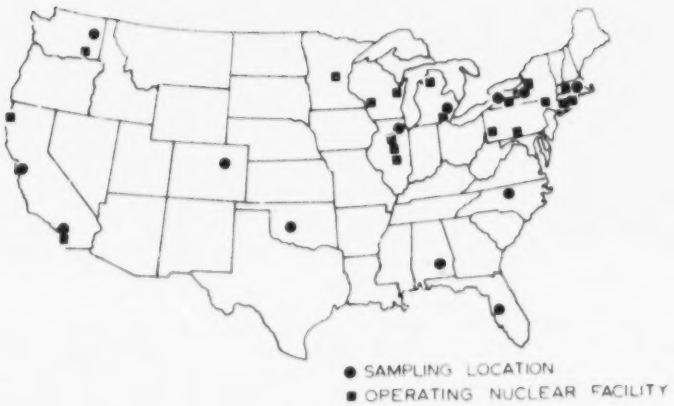

FIG. 1. Nuclear facilities in operation and sample collection sites.

TABLE I. CONCENTRATIONS OF ^{85}Kr IN AIR (pCi/m^3 of air (STP))

Location	Sample collection period			
	July-Sep. 1970	Oct.-Dec. 1970	Jan.-Mar. 1971	Apr.-Jun. 1971
Chicago, Ill.	-	-	-	14.7
Camden, N.J.	-	15.8	15.1	-
Denver, Colo.	16.2	17.0	-	-
Detroit, Mich.	-	14.4	15.0	-
Framingham, Mass.	-	15.2	15.2	-
Greensboro, N.C.	-	15.7	14.7	14.4
Las Vegas, Nev.	-	-	-	14.2
Liverpool, N.Y.[a]	-	-	46.8[a]	-
Long Beach, Calif.	-	14.8	14.4	-
Montgomery, Ala.	-	-	15.8	15.2
Oakland, Calif.	-	-	-	14.6
Oklahoma City, Okla.	-	-	14.9	-
Spokane, Wash.	-	-	-	20.6
Tonawanda, N.Y.	-	-	14.8	15.7
Tampa, Fla.	-	-	14.4	-

[a] Determined invalid due to collection technique.

capability was developed and reported by Cummings et al. [7] in these proceedings. The analyses had to be rapid enough to prepare at least one sample per day in order to produce a representative number of analyses.

Second, samples had to be representative of public breathing air. Cylinders containing about six cubic meters of compressed air were bought from commercial suppliers who furnished sample location and date of compression.

Third, sampling locations were selected in a manner which would allow the objectives of the study to be accomplished. Ideally locations would represent most of the major population centers and include sites with either a current or projected source for elevated ^{85}Kr concentrations. A few supposedly unaffected sites should also be included for comparison between normal and abnormal conditions.

Fourth, the sampling frequency had to provide sufficient samples to reflect any seasonal variations as well as national trends. Initially a quarterly sampling frequency was selected.

3. RESULTS

Current operating nuclear reactors and fuel reprocessing plants and locations of the sampling sites are indicated in Figure 1. The current analytical results are presented in Table I.

4. DISCUSSION

Some trouble was encountered in finding commercial suppliers in many of the selected cities. This caused a delay in acquiring many of the initial samples. Once this was accomplished, network sample collection became routine. Attention was also given to the method the supplier used to get compressed air. A sample from Liverpool, New York, was declared invalid because the supplier mixed liquid oxygen and liquid nitrogen to "make" air. The krypton is believed to have been concentrated in the liquid oxygen.

Data presented in Table I represent analyses of at least two aliquots of air from each cylinder. The mean value of 60 analyses was as follows:

Mean	Variance	Standard Deviation
15.3 pCi/m^3	1.78 $\left(\frac{pCi}{m^3}\right)^2$	$\pm$ 1.33 pCi/m^3

One sampling site, Spokane, Washington, exceeds the mean value by greater than two standard deviations. This has occurred for four aliquots from two separate samples. This phenomenon has yet to be clarified in detail although Spokane is reasonably close to the nuclear facilities at Hanford. Sax [8] has observed similar effects in New York at Springville and Albany from a Nuclear Facilities Service fuel reprocessing plant in that vicinity.

Figure 2 shows a comparison of the data from this study and the projections by Coleman and Liberace [6]. The current average value is in the upper range of values forecast by Coleman and Liberace.

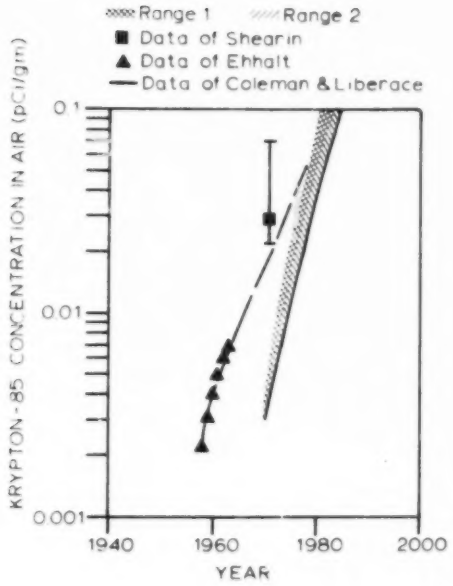

FIG.2. Study data versus predictions [6].
Range 1 = Upper range for 75% release in northern hemisphere.
Range 2 = Range of values for dilution in entire atmosphere.

Current annual dose rates calculated on the basis of an infinite cloud according to ICRP Publication No. II (9), are 30 μrem/a for the average concentration of 15.3 pCi/m^3. This average is about twice the value reported by Ehhalt et al. for 1963 [4].

5. CONCLUSION

This study indicates it is feasible to set up and operate a national surveillance network for monitoring the concentrations of krypton-85 in the environment. Although the current contribution to the population dose is insignificant compared to natural background sources, the rate of increase is significant and does warrant consideration. Particular attention should be paid to areas near fuel reprocessing plants where significantly higher concentrations of ^{85}Kr can occur.

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DISCUSSION

C.-O. WIDELL: Have you made any special investigation of the process by which krypton concentrates in liquid oxygen?

R.L. SHEARIN: No, but in addition to the work described in the paper, we have assayed pure O_2 and found unusually high concentrations. We have yet to arrive at a proper understanding of this effect.

A. ZIV: What is the lowest sensitivity attainable by your method for ^{85}K measurement?

R.L. SHEARIN: Our minimum sensitivity is less than 5 pCi/m^3, most likely as low as 2 pCi/m^3. At 14 pCi/m^3 we have a 7% error at the 95% confidence level.

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